If you are using a printed copy of this procedure, and not the on-screen version, then you <u>MUST</u> make sure the dates at the bottom of the printed copy and the on-screen version match.

The on-screen version of the Collider-Accelerator Department Procedure is the Official Version. Hard copies of all signed, official, C-A Operating Procedures are available by contacting the ESSHQ Procedures Coordinator, Bldg. 911A

C-A OPERATIONS PROCEDURES MANUAL

9.1.15.a BLIP Target and Canning Record

		Hand Processed Changes				
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	_	Collider-Accelerator Department Chairman Dat				

D. Beavis, S. Smith

Note 1: Uncontrolled copy printed from electronic master that is valid at time of printing. Always check that you have the latest revision of this document before use.

Note2: Leave no blanks. Indicate 'Not Applicable (N/A)', where appropriate.

Title and Preparer

Production of High Specific Activity ¹⁸⁶Re from Tungsten Metal

for Research and Clinical Applications

By Suzanne Smith and Matt Gott

	Instructions	
	Description	Page No.
1.	Overview [short summary of purpose of experiment; name of principle investigator and researcher involved]	4
2.	Target Material and Properties – [Provide physical properties of each component/material to be irradiated]	5
3.	Target Canning Process – [provide images or drawings and reference the OPM procedures for closing and opening of target can]	6
4.	Beam Characteristics [define required beam on target and total current required]	7
5.	7	
	a. Procedure for Irradiation of Target Material BLIP [Summarize steps for experiment including specialist and contact hours required for task]	7
	b. Target Array [Define proposed target array for box 1 and box 2 including SRIM calculated entry and exit energy for each layer. Provide physical dimension of degraders, target can, materials and water gaps]	7
	c. Thermal Analysis of Target Materials and Target Can [Provide full description of data provide to specialist for calculations and any assumption made on material for calculations]	7

	d. Transport and Processing at TPL [Provide full description of task involved and responsible persons and contact hours required]	7
	e. Disposal of waste. [describe waste to be generated and how it will be disposed of]	7
6.	Activation Analysis of Target Material and Can [Provide full list of radionuclide produced and quantities, references used for calculations, as well as decay profiles if the dose rates exceed limit for removal from BLIP hot-cell. Ensure Health Physics has reviewed data and confirms decay requirement if they are dose related. Attach analyses if any.]	8
7.	Expected Dose Rate (e.g., R/h at 1 m) [provide expected dose rate using <i>Microshield or equivalent</i> calculations for the combined and separate target and can irradiated. Provide expected dose rate at EOB at BLIP and expected dose rate when delivered to TPL]	8
8.	Additional Safety Requirements [address hazardous issues related to volatiles and or corrosive materials used and any additional equipment required for this experiment; hazardous materials information must be submitted to the C-AD ESSHQ Division Head for concurrence]	8
9.	Special Operating Instructions and List of References or Supporting Documents	9
10.	Appendix [provide additional support information as required]	10-16

1. Overview

Production of High Specific Activity ¹⁸⁶Re from W for Research and Clinical Applications.

Principal Investigator: Suzanne V Smith (Production Manager, MIRP BNL) **Researcher:** Matthew D Gott (Visiting PhD Student, Univ of Missouri)

Background: The irradiation described in this safety submission is part of planned research program entitled "Targetry Development and Production of ¹⁸⁶Re and ⁷²As for Research and Radiopharmaceutical Applications." It is a collaborative research program between Brookhaven and the University of Missouri. Matthew Gott received funding from the DOE Office of Science to visit Brookhaven to work on this project with Suzanne Smith.

The work described here is to specifically address the production of high specific activity 186 Re. 186 Re is a β^{-} emitter with favorable nuclear properties that makes it an ideal therapeutic surrogate for 99m Tc.

This safety submission outlines the issues related to the production of 186 Re from a tungsten metal foil. The main nuclear reaction of interest in this irradiation is 186 W(p, n) 186 Re. The irradiation will involve exposing the tungsten metal foil at an average current of 115 μ A for 1 hour.

Once irradiated, the target (containing the tungsten disc) will be transported to Target Processing Lab (TPL) in the transport cask, where it will be disassembled in a hot cell. The tungsten foil will then be transferred to one of radiochemistry laboratories (either 57a or 59) to test an established separation method developed by Matt Gott (at MU).

2. Target Material and Properties								
Target Name: Tungsten	Foil		T arget & Can ign unique no	_	x)			
		Targe	et Material P	roperties				
Purity or Grade	Tung	gsten Foil (9	99.95%) (See	Appendi	x 1 for Analys	is)		
Chemical Formula	W							
Physical Characteristics at 70 °F or 21 °C	Solid	ł						
Physical Form	Foil		Yes		Powder	No		
	Diame (inche		1 inch / 25	mm	Pressed (Torr)	N/A		
Elements (%)	W	(99.95%)						
Melting Point	34	422	°C		6192	°F		
Boiling Point	59	930	°C		10706	°F		
Thermal Conductivity	173	173 W.m ⁻¹ .K ⁻¹ Temperature dependence			(if available)			
Density	19.25	5			g/cm ³			
Specific Heat	133				J/kg.K			
		Target Mat	erial Reactio	ns / Prope	rties			
Does the Target material	Alun	ninum	No	Aiı	r No	CO ₂	No	
react with any of the following?	H	H ₂ O	No	Lea	d No	Zinc	No	
	Incor	nel 600	No	S/Ste	el No	Copper	No	
		Cannii	ng Material F	roperties				
Chemical Formula		Aluminum (6061)						
Can Wall Thickness (inche	s/mm)	mm) 0.02 inch / 0.508 mm						
Can Dimensions (inches/mm)			neter 2.75 in /	69.85 mn		0.48 in / 12.19	mm	
Melting Point		660.32		°C	1220	°F		
Thermal Conductivity		205 W.m	i ⁻¹ .K ⁻¹	Tempera (if availa	erature dependence ilable)			
Density		2.698		g/cm ³				
Specific Heat			910		J/kg.K			

Tungsten metal foil (99.95%) was purchased from Alfa Aesar and will be cut to the appropriate diameter (2.54 cm/1 inch). The tungsten foil will be sealed in the screw bolted aluminum can (figure 1 Appendix 4) within in a glove box under helium gas. The sealed aluminum can (containing the tungsten) will be leak tested for helium prior to placing it within the target assembly box behind the ⁸²Sr target array. A new degrader will be substituted for the current vacuum degrader box to ensure there is room within the box for the target. Selected degraders will be positioned within the target array to ensure optimum proton entry energy into the tungsten foil and to minimize the activation of water (see Appendix 2 and 3 for old and new target array).

3. Target Canning Process

	4. Beam							
Maximum Instantaneous Current Desired	40000	μА						
Average Current Desired	115	μА						
Total Integrated Current Desired	115	μA - hrs						
Maximum Proton Energy on Target Material	~5	MeV						

5. Experiment

- **5.a Procedure for irradiation of target material in BLIP:** The linac beam will be tuned to 116.8 MeV and the target array described in C-A-OPM 19.17.20a (See Appendix 2) will be modified behind the second RbCl target to ensure entry proton energy into the tungsten target will be approximately 5 MeV (See Appendix 3). The target will be exposed to an **average current of 115 \muA for 1 hour.** The irradiated tungsten target will be removed from beam and will be checked for radiation dose prior to transport to TPL in the transport cask (as per RWP: BLIP-15-007). If the radiation dose from the target is too high to comply with the specified RWP, it will be allowed to decay in the BLIP hot cell for 24 hours prior to transport to the TPL hot cell.
- **5.b** Target array in Box 1 and Box 2: The proton entry energy into the target assembly box will be set to 117 MeV. For the new target array, entry and exit proton energies for each degrader after the second RbCl target and into tungsten foil were calculated using TRIM and SRIM (see Appendix 3).
- Thermal analysis of target material and target can (attach analyses if any): The steady state thermal analysis of the irradiated tungsten foil and aluminum can was conducted using ANSYS software. Under the experimental conditions described, the maximum achievable temperature for the tungsten foil and the aluminum can were found to be 316°C and 228°C, respectively. These are well within the melting point thresholds for each component (i.e. 3422 °C for W and 660 °C). Details of the thermal analysis are given in Appendix 5.
- **5.d Transport of irradiated tungsten foil:** As described in section **5.a** above.
- **5.e Disposal of waste:** The aluminum can will be disposed with usual radioactive waste stream of the MIRP program. The liquid waste will be disposed in D tank system as described in OPM 19.30.3.

6. Activation Analysis of Target Material and Can

A conservative activation analysis was conducted using the increased isotope production rate at 10 MeV for the full irradiation period (i.e. 115 μ A for 1 hour). The list of radionuclides produced from irradiating tungsten metal and their respective yield is given in Appendix 6. For the tungsten foil, the primary isotopes that will be present are 182a,182b,186 Re with smaller quantities of 183,184 Re, and 183 Ta.

Decay Requirements

If necessary, the irradiated target will be allowed to decay for 24 hours prior to transport to hot cells in the TPL (as per RWP: BLIP-15-007). More specifically, for transport of radioactive sources from BLIP to TPL, the dose in the general area must be ≤ 2 mR/h and the dose on contact must be ≤ 100 mR/h.

The irradiated target will be opened in a hot cell in the TPL and the tungsten foil will be removed for further chemical processing and analysis in Lab 57a. According to RWP: RRPL-14-024 for Lab 57A, the dose in the general area must be ≤ 0.2 mR/h and ≤ 5 mR/h on contact, while the dose in the work area (fume hood) must be ≤ 100 mR/h at 30 cm from the source and ≤ 15 mR/h to the whole body.

7. Expected Dose Rate

The dose rate from the irradiated tungsten foil was calculated using Microshield. They are based on the maximum quantities of radioisotopes produced using an average current of 115 μ A over 1 hour. (See 15 MeV data in Appendix 6)

Shielding Configuration		Dose at EOB (mR/h)	Dose after 24 hour decay (mR/h)
Tungsten Foil inside Hot Cell	minimum	0.024	0.012
	maximum	0.095	0.052

8. Additional Safety Requirements

NIL

	9. Special Operating Instructions					
	3. Special Operating Instructions					
	Supporting Documentation					
References	 Lapi et al.; Appl. Radiat. Isot., 2007, 65, 345-349. Tarkanyi et al.; Nucl. Instr. Meth. Phys. B, 2006, 252, 160-174. Description of processing of tungsten target attached in paper entitled Radiochemical Study of Re/W Adsorption Behaviour on a Strongly Basic Anion Exchange Resin, Radiochemica Acta. 2014, 102(4), 325-332. 					
Calculations	Heat calculations checked by Chris Cullen, CAD, BNL.					

Appendix 1.

Certificate of Analysis for Tungsten Metal Foil from Alfa Aesar

Product No.: 10416

Product: Tungsten Foil, 99.95% (metals basis)

Lot No.: N09B003

	W		> 99.95 %
Al	< 0.002 %	Mn	< 0.002 %
С	< 0.005 %	Ni	< 0.003 %
Ca	< 0.003 %	Pb	< 0.002 %
Cr	< 0.002 %	Si	< 0.002 %
Cu	< 0.002 %	Sn	< 0.002 %
Fe	< 0.003 %	Ti	< 0.002 %
Mg	< 0.002 %		

Appendix 2.

Current Sr-82 Target Array from C-A-OPM 19.17.20a

Layer	Layer	Layer	Thi	ckness	Entry Proton	Exit Proton
No		Material	Inch	mm	Energy (MeV)	Energy (MeV)
	Window 1	Beryllium	0.012	0.305	116.8	116.5
	Window 2	AlBeMet	0.012	0.305	116.5	116.2
	Beam Line Window	Stainless Steel	0.031	0.787	116.2	113.3
	Water Gap	Water	0.106	2.692	113.3	111.5
	Box Window	Stainless Steel	0.020	0.508	111.5	109.6
1	Water Gap	Water	0.200	5.080	109.6	106.1
2	SS Degrader	Stainless Steel	0.029	0.737	106.1	103.7
3	Vacuum Box	Vacuum Box	0.596	15.140	103.7	103.7
4	SS Degrader	Stainless Steel	0.029	0.737	103.7	100.2
5	Water Gap	Water	0.200	5.080	100.2	96.5
6	Target Front	Inconel 600	0.012	0.305	96.5	95.2
7	RbCl Target 1	RbCl	0.646	16.400	95.2	73.7
8	Target Back	Inconel 600	0.012	0.305	73.7	72.1
9	Water	Water	0.200	5.080	72.1	67.2
10	Target Front	Inconel 600	0.012	0.305	67.2	65.5
11	RbCl Target 2	RbCl	0.500	12.700	65.5	42.7
12	Target Back	Inconel 600	0.012	0.305	42.7	40.2
13	Water Gap	Water	0.200	5.080	40.2	32.2
14	Cu Degrader	Copper	0.220	5.590	32.2	STOP
15	Box Window	Stainless Steel	0.020	0.508	NIL	

A modification to the standard C-A-OPM 19.17.20a ⁸²Sr target array is being proposed in order to conduct the tungsten foil irradiations. The changes include:

- 1) Substituting the vacuum box with a 0.58" stainless steel degrader to achieve the appropriate beam degradation specific for needs for ⁸²Sr production. This change will provide for more space in the target assembly box that will allow positioning of the aluminum can target behind the RbCl targets.
- 2) Replacing the copper degrader (originally at the back of the RbCl target) with a new aluminum degrader.
- 3) The new target array is illustrated in Appendix 3.

Appendix 3.

New Target Array for Irradiation of Tungsten Foil in Al Can (Sr-82_W_02)

Entry and Exit Proton Energies for Target determined using TRIM and SRIM

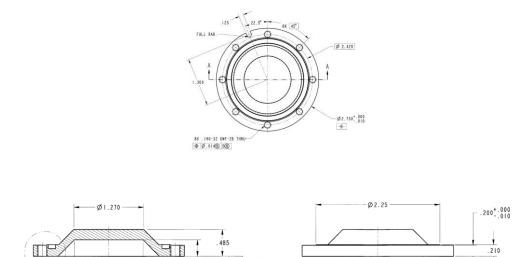
Layer	Layer	Layer		ckness	Entry	Exit Proton
No.		Material	Inch	mm	Proton Energy (MeV)	Energy (MeV)
	Window 1	Beryllium	0.012	0.305	116.8	116.5
	Window 2	AlBeMet	0.012	0.305	116.5	116.2
	Beam Line	Stainless	0.031	0.787	116.2	113.3
	Water Gap	Water	0.106	2.692	113.3	111.5
	Box Window	Stainless	0.020	0.508	111.5	109.6
1	Water Gap	Water	0.200	5.080	109.6	106.1
2	SS Degrader	Stainless	0.058	1.473	106.1	100.2
3	Water Gap	Water	0.200	5.080	100.2	96.5
4	Target Front	Inconel 600	0.012	0.305	96.5	95.2
5	RbCl Target 1	RbCl	0.646	16.400	95.2	73.7
6	Target Back	Inconel 600	0.012	0.305	73.7	72.1
7	Water	Water	0.200	5.080	72.1	67.2
8	Target Front	Inconel 600	0.012	0.305	67.2	65.5
9	RbCl Target 2	RbCl	0.500	12.700	65.5	42.7
10	Target Back	Inconel 600	0.012	0.305	42.7	40.2
11	Water Gap	Water	0.200	5.080	40.2	31.7
12	Al Degrader	Aluminum	0.068	1.720	31.7	24.8
13	Water Gap	Water	0.200	5.080	24.8	9.8
14	Target Front	Aluminium	0.020	0.508	9.8	4.4
15	W Metal Target	Tungsten	0.002	0.050	4.4	1.6
16	Target Back	Aluminium	0.178	4.521	1.6	NIL
17	Water Gap	Water	0.298	7.569	NIL	NIL
18	Water Gap	Water	0.100	2.540	NIL	NIL
19	Al Degrader	Aluminum	0.056	1.422	NIL	NIL
	Box Window	Stainless	0.020	0.508	NIL	NIL

Sr-82_W_02 Target Array Updated with Data from Short Tungsten Foil Irradiations

10	Target Back	Inconel 600	0.012	0.305	43.7	41.2
11	Water Gap	Water	0.200	5.080	41.2	33.0
12	Al Degrader	Aluminum	0.068	1.720	33.0	26.2
13	Water Gap	Water	0.200	5.080	26.2	12.5
14	Target Front	Aluminium	0.020	0.508	12.5	8.3
15	W Metal Target	Tungsten	0.002	0.050	8.3	6.4
16	Target Back	Aluminium	0.178	4.521	6.4	NIL
17	Water Gap	Water	0.298	7.569	NIL	NIL
18	Water Gap	Water	0.100	2.540	NIL	NIL
19	Al Degrader	Aluminum	0.056	1.422	NIL	NIL
	Box Window	Stainless	0.020	0.508	NIL	NIL

Appendix 4.

Drawing of Aluminum Screw Bolted Target Can and Loading Procedure.



Target Can – Aluminum Bolted (extract from D25-M-3186)

Procedure for loading of target into Aluminum Bolted Can

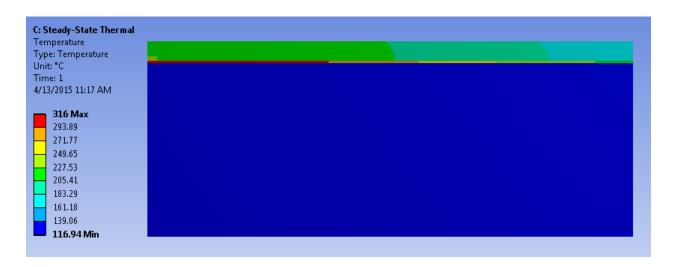
Aluminum Bolted Can – modification for target.

- The aluminum capsule consists of two parts referred to as upper half (blueprint D25-M-3188), normally positioned upstream beam entry and lower half (blueprint D25-M-3187) positioned downstream (beam exit) that are secured together with eight Allen screws located at the periphery of the target.
- 2. The water-tight seal is ensured by a c-ring
- 3. In the center of the lower half there is a well, which is designed to hold target material. It is machined specifically for each target. In this study, 0.002 inch depth for the tungsten foil with a diameter of 1.0 inch.
- 4. The tailoring of the target well is to ensure optimal thermal conductivity.

Loading of Target Material into Aluminum Bolted Can.

- a. Label the upper part placing the letter stamps between the screw holes
- b. Cut tungsten foil to 1.0-inch diameter circles ensuring they fit the machined well of the aluminum bolted can.
- c. Clean the well of target can and c-ring with acetone and leave to air dry.
- d. Place the c-ring in the grove of the lower half of the capsule
- e. Place the tungsten foil in the well.
- f. Position the upper half of the capsule on top of the lower half and align the indexing slits
- g. Insert eight Allen screws into the holes individually secure in the following order: position 12-6-9-3-11-4-1-7 o'clock
- h. Create a target card as per C-A-OPM-ATT 19.3.6.2.a
- i. Transfer loaded target can to BLIP operator for loading into target assembly.

Appendix 5.
Solid State Thermal Analysis



Calculation conditions: 2D Axi-symmetric model, 0.002" thick tungsten foil in a bolted aluminum can, Steady-state analysis, 2000 W/m².°C thermal contact resistance between each layer, 5000 W/m².°C water cooling convection on exterior, Gaussian internal heat generation for all bodies.

Experimental conditions used for the calculations: 116.8 MeV protons (entry to target assembly box), average current of 115 μ A for 1 hour assumed 60% beam on target. Total energy deposited in the tungsten foil is 1.9 MeV as given in target array in Appendix 3. Analysis shows the maximum achievable temperature for the tungsten foil and the aluminum can were found to be 316° C and 228° C, respectively. These values are well below the respective melting points.

Appendix 6.

Literature Cross Sections and Calculated Activities for Anticipated Isotopes for Os Disc

Radioisotopes	σ (mb) at 10 MeV	t _{1/2} of product	Calculated Activity (mCi)	Calculated Activity (mCi) at Higher and Lower Average Proton Energies		
	10 1016	(hours)	1 hour irradiation	5 MeV	15 MeV	
Re-181	0.00	19.9	0.00	0.00	100.10	
Re-182 (a)	8.02	12.7	9.49	0.00	20.32	
Re-182 (b)	86.53	64.0	4.34	1.54	24.47	
Re-183	46.25	1680	0.19	0.00	1.12	
Re-184	59.74	849.6	0.50	0.00	0.15	
Re-184m	0.00	4056	0.00	0.00	0.00	
Re-186	62.26	89.23	4.91	1.51	1.58	
Ta-177	0.00	56.56	0.00	0.00	6.69	
Ta-183	1.14	122.4	0.07	0.00	0.92	

Appendix 7.

Identifiable Gamma Energies for Anticipated Isotopes for W Foil

Isotope	Half-Life	Gammas (keV)	Intensity (%)
Re-181	19.9 h	365.5	56
Re-182	64.0 h	169.2	11.4
		1121.4	22.1
Re-182m	12.7 h	1121.4	22.1
Re-183	70.0 d	162.3	23.2
Re-184	35.4 d	792.1	37.7
Re-184m	169 d	104.7	13.6
		792.1	3.69
Re-186	3.718 d	137.2	9.47
Ta-177	56.6 h	112.9	7.2
Ta-183	5.1 d	246.24	27
		162.3	4.9
Decay Daughter:			
W-181	121.2 d	136.3	0.03